Role of Temperature and Substrate in the Scalable and Uniform Deposition of MoS_2 by Mo Thin Films Sulfurization

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Two-dimensional (2D) materials represent nowadays the new frontiers of semiconductor technology [1]. At the one side, great effort has been done in research applied to graphene, producing new technology that will rebuild many industrial sectors. On the other side, non-graphene 2D materials, like transition metal dichalcogenides (TMDs), are highly attractive because they offer complementary properties to graphene [2], but still lack a large-scale production method for high quality and well controlled layers.

MoS_2, one of the most studied TMD materials, has been produced by using many techniques, but the deposition through chemical methods, i.e. based on the use of materials such as Mo or MoO_3 and then the reaction with sulfur, already demonstrates to fit better the stringent requirements of lateral uniformity on the centimeter scale, vertical scalability and structural optimization as function of the growth parameters [3].

However, many details about the chemistry of the reaction between Mo precursor and sulfur needs further clarification, in view of a large-area production [4]. In particular, it is not clear what are the roles of: *i)* the materials used as precursors, *ii)* the substrate type and *iii)* the deposition temperature, which drives not only the Mo-S reaction, but also other undesired ones.

This work is focused on the study of some of these fundamental parameters in the growth of MoS_2 on SiO_2/Si substrates by chemical reaction between a Mo pre-deposited thin film precursor (TFP) and sulfur (figure 1*(a)*). We demonstrate that the control of the Mo film thickness allows the accurate control of the MoS_2 thickness. This methodology leads to MoS_2 nanosheets with a tunable number of layers and uniformly extended throughout the whole area of the supporting substrates (figure 1*(b)*). In this type of process, the role of the growth temperature and of the substrate on the structural properties of the layers turns out to be dramatically important.

Various techniques are employed to characterize the MoS_2 layers. In particular, Raman spectroscopy is the key-probe used for single- and multi-layers identification and for the validation of the high structural quality of the grown material (see figure 1*(b)*). At the same time, fundamental information about the chemical details, the morphology, and the structural quality is also drawn from X-ray photoelectron microscopy (XPS), atomic force microscopy (AFM), and photoluminescence (PL).

The temperatures used for the present study are 500, 750, and 1000 °C. All the grown films show granular morphology by AFM (typical grain size: 20-50 nm). Best results in terms of layer quality are obtained at the highest sulfurization temperature and detailed analysis of Raman spectra collected from MoS_2 quadrilayers demonstrates that the layer features are comparable to those of exfoliated MoS_2 samples.

We further demonstrate that SiO_2/Si wafers bring intrinsic limitation as a substrate for the MoS_2 growth at high deposition temperatures (1000 °C), because of the activation of silicon diffusion from the substrate and of the chemical reaction between Si and S with the formation of the SiS_2 byproduct [5].

Our study supplies some fundamental information on the sulfurization process of Mo pre-deposited thin films, clarifying the role of temperature in yielding high quality materials and putting a warning on the use of silicon substrates for this type of process [6].

References

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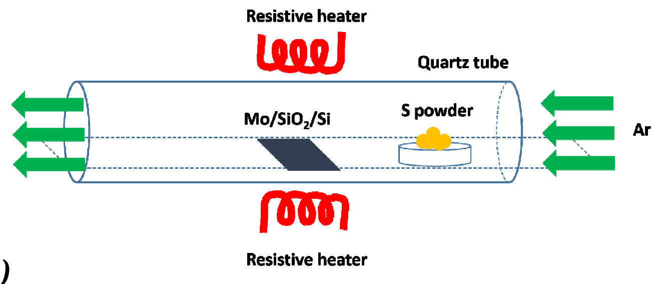
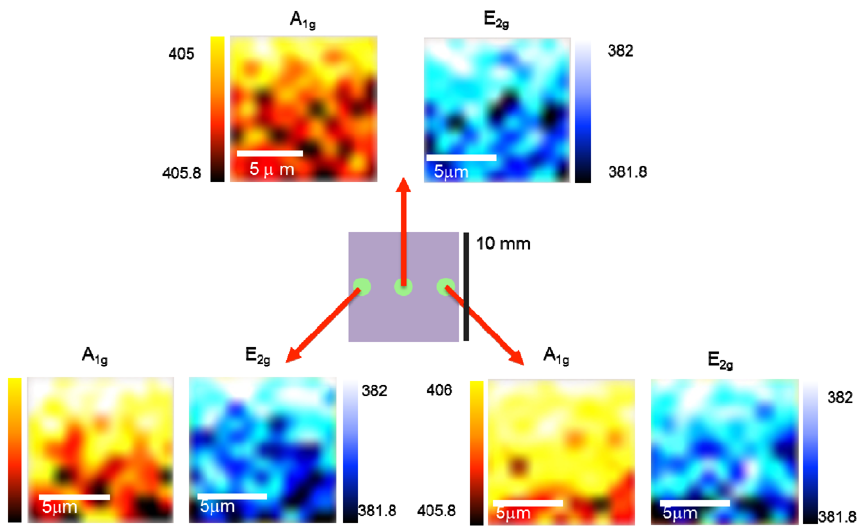
[6] This research was partially supported by US-Army Contract W911NF-14-1-0612 with CNR-IMEM and CNR-IMM.



a)



b)

Figure 1. ***(a)*** Sketch of the sulfurization method.
(b) Raman maps of a 4-layers MoS_2 film grown at 1000 °C. In particular, the maps report the value of the A_{1g} (yellow-red colored) and E_{2g} peaks (blue colored) positions in an area of 10x10 μm^2, testing three points of the sample. The variation of $\Delta\omega$ inside the area has values comprised between 0.7 and 1.3 cm^{-1}, compatible with the Raman instrument sensitivity.